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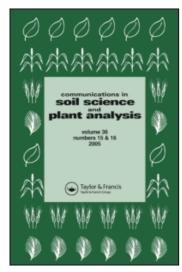
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Impact of Time to First Rainfall Event on Greenhouse Gas Emissions Following Manure Applications

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Use of inorganic fertilizers and manures are known to result in the release of greenhouse gases (GHG) to the atmosphere, and rainfall events can also increase GHG emissions from soils. The objective of this study was to examine how the time between fertilizer or manure application and the first rainfall event affects carbon dioxide (CO_2), nitrous oxide (N_2O), and methane (CH_4) fluxes. Swine manure, poultry litter, and urea were surface applied to plots. Rainfall was simulated 1, 4, 8, 15, or 29 days after application. Gas fluxes were determined before and after each rainfall simulation. Postrain CO_2 fluxes were the greatest from poultry litter at 4 to 8 days after fertilization, and all fertilizer treatments produced similar N_2O emissions with a peak 4 days after fertilization. These data seem to indicate that if manures are applied during drier periods of the year, GHG emissions can be minimized, in addition to reducing nutrient runoff losses.

Keywords Carbon dioxide, methane, nitrous oxide, poultry litter, soil gas flux, swine manure, urea

Introduction

Greenhouse gas (GHG) emissions are presumed to induce global warming, and agriculture has been identified as a contributor to these gas accumulations in the atmosphere. Carbon dioxide is produced from soils as a result of aerobic respiration from microbes and plant roots and shoots. Nitrification and denitrification are the primary processes responsible for the conversion of nitrogen (N) in soil to nitrous oxide (N₂O) released to the atmosphere (Sahrawat and Keeney 1986). Methane (CH₄) can be released to the environment from rice production and animal manures; however, soils can also serve as a sink for CH₄ from the atmosphere (Sommer, Petersen, and Moller 2004; Willison et al. 1995).

Global estimates indicate that agriculture is the source for 35% of the N_2O (Kroeze, Mosier, and Bouwman 1999) and 30 to 50% of the CH₄ released to the atmosphere (Burke and Lashof 1990). Flessa et al. (2002) concluded that 60% of the total GHG emissions from animal operations were from N_2O and that 25% and 15% of the total emissions were from CH₄ and carbon dioxide (CO₂), respectively.

Greenhouse gases are also emitted from soils following the application of manure or fertilizers. Greater rates of swine manure application have been shown to increase CO₂

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emissions, and even soils fertilized with low levels of swine manure application demonstrated greater CO₂ emissions than soils fertilized with inorganic fertilizers (Rochette, Angers, and Cote 2000). Peak CO₂ emissions followed application of swine and dairy manure within 2 days to 2 weeks, respectively (Rochette, Angers, and Cote 2000; Calderon et al. 2004). In another study, following the application of dairy manure when CO₂ emissions were immediately greater, peak emissions were not observed for 30 to 80 days after application (Gregorich et al. 1998).

Nitrous oxide emissions have been shown to be greater from manures than from inorganic fertilizers, when applied at the same N rate, and peak fluxes from all sources occurred within 2 weeks of application (Akiyama and Tsuruta 2003). Following the addition of swine or dairy manure to soils, peak N_2O fluxes occurred within the first 2 weeks (Whalen 2000; Calderon et al. 2004). Li et al. (2002) compared N_2O emissions from cattle manure, ammonium sulfate and a controlled release fertilizer, and concluded that N_2O emissions were greatest from the controlled release fertilizer and least from the manure treatment. As in the other studies, peak N_2O emissions from all three fertilizer sources were within three weeks following fertilizer application (Li, Inubushi, and Sakamoto 2002). Another study compared N_2O emissions from ammonium nitrate (NH₄NO₃), slow-release N fertilizer, and urea and observed peak N_2O emissions within approximately 10 days of fertilization. The greatest emissions were from NH₄NO₃, whereas the least N_2O emissions were from the urea fertilizer (Maggiotto et al. 2000).

Soil is known to be a consumer and producer of atmospheric CH₄. Chan and Parkin (2001) observed the production of CH₄ from agricultural soils during a very wet year, whereas in a year with more normal precipitation, most agricultural soils consumed CH₄. However, while most agricultural soils were consuming CH₄, fertilization with swine manure resulted in the release of CH₄ to the atmosphere, primarily due to the release of dissolved CH₄ from the manure (Chan and Parkin 2001).

Little information has been gathered on how the time between fertilizer or manure application affects GHG emissions. However, CO₂ emissions from runoff have been studied, and peak emissions occur within the first few weeks after the runoff event (Jacinthe, Lal, and Kimble 2002). The objective of this study was to determine the impacts of the time between fertilizer or manure application and first rainfall event on GHG emissions.

Materials and Methods

Fifteen primary plots were built on an Octagon silt-loam soil (fine-loamy, mixed, mesic, Mollic Hapludalfs) at the Throckmorton Purdue Agricultural Research Center, near Lafayette, Ind. Each plot consisted of four subplots measuring 0.75 m wide and 2 m long. Plots and subplots were hydrologically isolated from the surrounding soil by inserting 0.2-m-wide 14-gauge galvanized sheet metal borders approximately 0.1 m into the soil. Approximately 0.2 m upslope of each subplot, an anchor for GHG emission chambers was inserted into the soil. Chamber anchors, which measured $73.7 \times 35.4 \times 12.0$ cm, were inserted 10 cm into the soil. A U-shaped channel, measuring 1.8 cm wide by 1.9 cm deep, was welded to the outer edge of the chamber anchor. During flux measurements, the U-shaped channel was filled with water, and the chamber lid, measuring $75.8 \times 38 \times 13$ cm, was placed into the U-shaped channel, forming an airtight seal (Figure 1). Flux measurements are described in further detail later.

For this study, treatments were (1) inorganic fertilizer, (2) poultry litter, (3) swine manure, and (4) unfertilized control. Swine manure was applied at a rate equivalent to 27,000 L ha⁻¹, and poultry litter was applied at a rate equivalent to 4,200 kg ha⁻¹. There



Figure 1. Photograph showing the gas flux chamber and chamber anchor used to measure greenhouse gas fluxes. The chamber anchor is installed semipermanently in the soil. The gas flux chamber fits into the U-shaped channel and is used only during the flux measurement period.

were three replications of each treatment and each time combination. Inorganic fertilizer was applied as triple superphosphate at 30 kg P ha⁻¹ and urea at 100 kg N ha⁻¹. Treatments were randomly assigned to subplots, with manure or inorganic fertilizer applied to the area of the assigned subplot, and the corresponding area within the GHG anchor.

Rainfall simulations were carried out 1, 3, 8, 15, and 29 days after fertilization at an intensity of 100 mm h⁻¹ for approximately 1 h, 15 min. Prior to each rainfall, and 24 h after each rainfall, GHG fluxes were calculated for each subplot. To determine fluxes of CO₂, N₂O, and CH₄, a vented aluminum flux chamber was placed in the channel around the anchor. Prior to placement of the chamber, water was poured into the channel to assure an airtight seal. Individual gas samples were withdrawn from the chamber at 0, 5, 10, and 15 min following chamber placement, using a 30-mL syringe. From this syringe, 20 mL of the gas sample was injected into a 10-mL evacuated vial, which was overpressurized so that if any gas leaks existed, there would not be contamination from ambient air. Upon arrival at the laboratory, gas samples were analyzed for CO₂, N₂O, and CH₄ using a Varian model 3900 gas chromatograph equipped with a thermal conductivity detector (TCD), electron capture detector (ECD), and flame ionization detector (FID). Gas fluxes were calculated using the ideal gas law.

Carbon dioxide equivalents (eCO₂) were calculated from the flux of each gas during the sampling periods. Fluxes for CO₂, N₂O, CH₄, and eCO₂ were statistically analyzed using analysis of variance procedures (ANOVA) in SAS version 8.0 (SAS Institute, Cary, N.C.). Fisher's protected least significant difference (LSD) procedure was used for separation of means, with an a priori level set at P < 0.05.

Results and Discussion

Meteorological data for the study period are presented in Figure 2. Natural rainfall did occur on 14 of the 29 days of this field study. During natural rainfall events, plots were covered with plastic to reduce the potential for runoff losses and minimize potential impacts from the rainfall on gas emissions.

Gas Emissions before Rain

CO₂ fluxes before rain were approximately 7,700 μ g CO₂ m⁻² h⁻¹ for all treatments except the swine manure (11,000 μ g CO₂ m⁻² h⁻¹) 1 day after fertilization (Figure 3A). A similar observation was made for N₂O fluxes calculated for the same period, with swine manure emitting approximately 160% more N₂O than the other treatments (Figure 3B). Methane fluxes were positive for swine manure (4.6 μ g CH₄ m⁻² h⁻¹) and inorganic fertilizer (3.1 μ g CH₄ m⁻² h⁻¹) treatments, whereas CH₄ sequestration was occurring for the other treatments 1 day after fertilization (Figure 3C).

There was a slight decrease in CO_2 fluxes between days 1 and 4 for all treatments except the poultry litter fertilization. Peak CO_2 fluxes occurred at day 8 for poultry litter (16,000 μ g CO_2 m⁻² h⁻¹), swine manure (15,000 μ g CO_2 m⁻² h⁻¹), and unfertilized (15,000 μ g CO_2 m⁻² h⁻¹) treatments, whereas peak CO_2 fluxes for the inorganic fertilizer treatment occurred at day 15 (14,000 μ g CO_2 m⁻² h⁻¹). The greatest difference between CO_2 fluxes from fertilized and unfertilized plots occurred at day 15. At the day 29 rainfall, the before-rain CO_2 fluxes from fertilization were lower than peak emissions and comparable to unfertilized control plots. The occurrence of peak CO_2 fluxes were intermediate

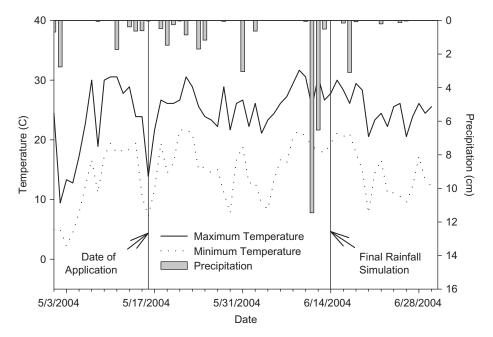


Figure 2. Daily maximum and minimum temperatures and precipitation for May and June 2004 during the study period. The vertical line at 16 May represents application of manure and fertilizer. The vertical line at 14 June represents final rainfall simulation for the experiment.

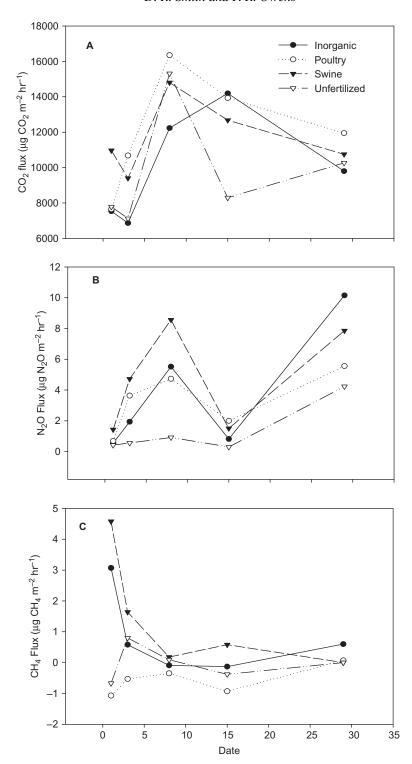


Figure 3. Mean fluxes of (A) CO_2 , (B) N_2O , and (C) CH_4 with time following application of inorganic fertilizer, poultry litter, and swine manure to pasture plots prior to simulated rainfall.

when compared to other studies, arising later than in studies by Rochette, Angers, and Cote (2000) and Calderon et al. (2004) and sooner than the 35 to 85 days observed by Gregorich et al. (1998).

There are two potential reasons for the differences between fertilized and unfertilized plots at days 15 and 29. The first possibility at day 15 is that fertilization increased respiration from soil microbes as well as the plants in the chambers, whereas the plants in the unfertilized plots were not growing as vigorously. The second issue involves heavy rains that occurred between days 26 and 28 (Figure 2). Although the plots were covered during this period, rainfall was most likely sufficient to increase the soil moisture content of the plots, resulting from interflow and return flow or seepage (see Haygarth et al. 2000). Increased soil moisture in the plot areas could result in a flush of CO₂ from a concomitant increase in biological (plant and microbial) activity.

N₂O fluxes before rain increased rapidly for all fertilized treatments up to day 8, at which time the relative difference in N₂O fluxes between fertilized and unfertilized plots was the greatest. Swine manure maintained the greatest N₂O fluxes for the first 8 days of the study, presumably because of the anaerobic nature of liquid swine manure, in which denitrifying bacteria would already be prevalent, or possibly because of the rapid ammonification of organic N followed by quick nitrification. These results contradict those of Akiyama and Tsuruta (2003), who found that N₂O emissions were the greatest from poultry manure. The swine manure in the current study was a liquid swine manure, whereas in the previous study the swine manure was dried and powdered following a 2-week aerobic composting process (Akiyama and Tsuruta 2003). Between day 15 and day 29, a marked increase in N₂O emissions was observed for all treatments, including the unfertilized plots. The low N₂O emissions at day 15 were likely due to the drier soil and thus predominantly aerobic respiration in the soil. Other studies have shown peak N₂O fluxes occurring within 2 weeks of fertilizer application (Maggiotto et al. 2000; Whalen 2000; Li, Inubushi, and Sakamoto 2002; Akiyama and Tsuruta 2003; Calderon et al. 2004). Peak N₂O emissions resulting from fertilizer treatments in the current study occurred on day 8 (Figure 3B), which concurs with results from the previous research. Fluxes of N₂O were, in some cases, greater on day 29 than on day 8; however, the unfertilized control plots increased by an order of magnitude between days 15 and 29 (0.3 and 4.2 μ g N₂O m⁻² h⁻¹, respectively). This would indicate that the N₂O flux increases observed on day 29 were most likely due to a reason other than the application of fertilizers to the plots.

At day 29, soils were wetter from the natural rainfall event (despite being covered during rainfall), and as a result, a greater proportion of microbial activity in the soil could have been carried out under anaerobic conditions, which is conducive to denitrification. The greatest N_2O fluxes at day 29 occurred from the plots fertilized with urea. This could be due to slower conversion from NH_3 released from urea through NH_4^+ to NO_2^- and then finally a gaseous loss of N_2O . Slower conversion from the inorganic N treatment would be expected, because the entirety of the N applied was in the urea form: there was neither a microbial pool nor a readily available C source applied with the inorganic fertilizers (such as the case with swine manure or poultry litter). Swine manure (4.6 μ g N_2O m⁻² h⁻¹) maintained the greatest mean N_2O flux prior to each rainfall event during the entire study, whereas the lowest mean N_2O flux for the study prior to each event was observed from the unfertilized plots (1.1 μ g N_2O m⁻² h⁻¹).

The greatest differences in CH₄ fluxes occurred 1 day after fertilizer application (Figure 3C). Plots fertilized with poultry manure ($-0.61~\mu g$ CH₄ m⁻² h⁻¹) maintained the lowest CH₄ fluxes for measurements before the rain, whereas swine manure (1.49 μg CH₄ m⁻² h⁻¹) contributed the greatest CH₄ fluxes throughout the study for measurements taken

Date	Unfertilized	Inorganic	Poultry	Swine
1	1.90 A	1.86 A	1.88 B	2.76 A
4	1.76 A	1.80 A	2.83 AB	2.61 A
8	3.74 A	3.35 A	4.27 A	4.19 A
15	2.01 A	3.47 A	3.49 AB	3.16 A
29	2.78 A	3.11 A	3.28 AB	3.17 A

Note. Numbers followed by the same capital letter within a column are not significantly different at P < 0.05.

prior to rainfall simulations. Results are similar to Chan and Parkin (2001), in which both positive and negative CH₄ fluxes were observed, with some of the greatest fluxes observed resulting from the release of dissolved CH₄ from land-applied swine manure.

Upon calculating the overall GHG emissions (as CO_2 equivalents or eCO_2) from the flux measurements taken prior to rainfall, there were no significant differences between fertilizer treatments (Table 1). Peak emissions for the study period did occur on day 8 (3.9 kg eCO_2 ha⁻¹ d⁻¹) with significantly greater emissions than the previous two measurements (2.1 kg eCO_2 ha⁻¹ d⁻¹ for day 1 and 2.3 kg eCO_2 ha⁻¹ d⁻¹ for day 3). These data would suggest that despite increased biological activity from inorganic and organic fertilizer additions, the overall emissions of GHGs (from CO_2 , N_2O , and CH_4) without rainfall are impacted less by fertilization than by other factors. However, one should be cautioned that calculation of eCO_2 takes into account the variability observed for all three gases and thus tends to have greater variability than any single gas flux. This statement is true only during dry periods, when rainfall would not affect the biological activity nor induce potentially anaerobic conditions at microsites in the soil.

Gas Emissions after Rain

Poultry litter supplied the greatest CO_2 emissions (15,000 μg CO_2 m⁻² h⁻¹) after a simulated rainfall occurring 1 day after fertilization; the emissions were more than 135% greater than the fluxes observed for the unfertilized plots after rainfall for this period (Figure 4A). The poultry litter treatment flux 1 day after fertilization following rainfall was also approximately twice as great as the flux observed prior to the rainfall simulation (Figure 3A). This suggests that the microbial populations in the poultry litter were able to start activity very quickly following the rainfall simulation. This could be a result of the low moisture content of the poultry litter (gravimetric water content of approximately 19%) and the high salt content of poultry litter. It would appear that respiration increased rapidly after the microbial populations were able to overcome their moisture limitation.

Following fertilization, CO_2 fluxes after the rain increased until day 8, after which fluxes declined (Figure 4A). The poultry litter treatment maintained the greatest CO_2 emissions during this period, with peak emissions of approximately 18,000 μ g CO_2 m⁻² h⁻¹ on days 4 and 8. Carbon dioxide fluxes from swine manure after the rainfall simulations were very similar to those observed for the unfertilized plots for the duration of the study. For the entire study period, poultry litter maintained the greatest CO_2 emissions after rainfall

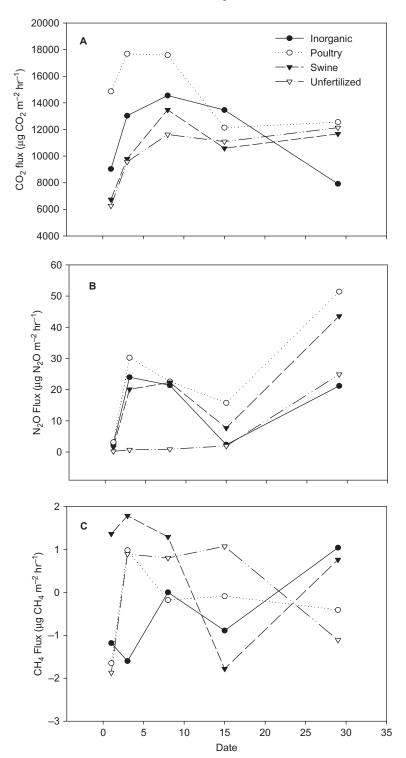


Figure 4. Mean fluxes of (A) CO_2 , (B) N_2O , and (C) CH_4 with time following application of inorganic fertilizer, poultry litter, and swine manure to pasture plots after simulated rainfall.

 $(15,000~\mu g~CO_2~m^{-2}~h^{-1}$ for poultry litter and a mean of 11,000 $\mu g~CO_2~m^{-2}~h^{-1}$ for the other treatments).

After the rainfall simulations, N2O fluxes 1 day after fertilization were an order of magnitude greater from the fertilized plots (mean flux of 2.4 µg N₂O m⁻² h⁻¹) than unfertilized plots (0.2 μg N₂O m⁻² h⁻¹; Figure 4B). The main effect of fertilizer treatment on N2O flux after rainfall was a significant increase resulting from manures compared to the unfertilized treatment (increases of 330% and 230% for poultry litter and swine manure, respectively). The main effects of time were that the greatest N₂O emissions occurred on day 29 and that fluxes from days 3 and 8 were significantly greater than fluxes observed on day 1. It would appear that N₂O fluxes were starting to decline by day 15. The N₂O fluxes were likely greater on day 29 as a result of the natural rainfall that occurred on days 26 through 28. As discussed previously, moisture likely entered the soil from which flux measurements were taken through interflow and return flow. This scenario is likely, as the storms that occurred during this 3-day period caused a flood with a 100-year return period in the local area. Given this information, when rainfall occurred on these plots after the excessive rainfall of the previous 3 days, the N₂O emissions after rainfall were most likely greater than they would have been without the excessive natural rainfall event.

The N_2O fluxes after rain were 50% less for the unfertilized treatment compared to the values observed prior to rainfall (Figure 4B). Increased N_2O fluxes after rainfall at day 1 were observed for poultry litter and inorganic fertilizer treatments compared to the beforerain values (increases of 350% and 390%, respectively). Relative increases in N_2O fluxes after rainfall were even greater for days 4 and 8 than day 1 compared to before-rain fluxes. This effect would be expected, as rainfall would induce anaerobic microsites in the soil. Denitrification would occur as O_2 depletion occurred at these microsites, yielding N_2O , which would then be released to the atmosphere.

Methane fluxes following rainfall were less than those observed prior to rainfall on day 1 (Figure 4C), and the only positive flux during this measurement period was from the swine manure treatment. There were no clear trends for CH₄ fluxes that occurred with time or due to fertilizer treatments.

The overall GHG emissions as measured by eCO_2 were greatest for the plots fertilized with poultry litter (5.4 kg eCO_2 ha⁻¹ d⁻¹ for poultry litter and 3.5 kg eCO_2 ha⁻¹ d⁻¹ for all others; Table 2). The temporal pattern observed with overall eCO_2 fluxes after rainfall was an increase up to day 8, followed by a decline at day 15. Although the greatest eCO_2 fluxes observed occurred at day 29, this most likely occurred because of the reasons stated previously; heavy natural rainfall amounts inundated the soil through interflow and return flow. From the analysis of the eCO_2 and eCO_2 data, it is apparent that aerobic respiration levels on day 29 were maintained at levels similar to day 15; however, the increases in eCO_2 occurred as a result of increased eCO_2 fluxes for all treatments.

Conclusions

Peak GHG emissions resulting from fertilizer or manure applications before and after rainfall simulations appear to have been the greatest within 2 weeks of application. Elevated gas emissions at day 29 during this study were most likely the result of an unusually high period of rainfall a few days prior to the final rainfall simulation. There were few differences in CO_2 fluxes prior to rainfall, but poultry litter induced the greatest CO_2 fluxes after the rainfall events. Swine manure produced the greatest N_2O emissions prior to rainfall because of its liquid nature, whereas poultry litter produced the greatest N_2O emissions

Table 2					
Effects of fertilization and time to first rainfall event on CO ₂ equivalent flux (kg eCO ₂					
ha ⁻¹ d ⁻¹) from soil cropped to pasture after rainfall simulations					

Date	Unfertilized	Inorganic	Poultry	Swine
1	1.51 B z	2.36 B z	3.79 B z	1.74 C z
4	2.35 AB y	4.91 AB yz	$6.50\mathrm{Az}$	3.85 ABC yz
8	2.86 AB y	5.08 A yz	$5.90\mathrm{AB}\mathrm{z}$	4.90 AB yz
15	$2.81\mathrm{AB}\mathrm{z}$	$3.40\mathrm{AB}\mathrm{z}$	$4.08\mathrm{B}\mathrm{z}$	3.10 BC z
29	$4.76\mathrm{A}\mathrm{yz}$	3.48 AB y	$6.83 \mathrm{Az}$	6.05 A yz

Note. Numbers followed by the same capital letter within a column are not significantly different at P < 0.05. Values followed by the same lowercase letter within a row are not significantly different at P < 0.05.

following rainfall. Fertilized and unfertilized soils acted as both a source and a sink for CH₄ during this study with no clear trend.

To minimize GHG emissions from fertilizer applications, producers should consider applying manure or fertilizers during drier periods of the year, when rainfall is not expected to occur for 1 week following the application. This information complements recommendations that suggest applying manures during drier periods to avoid nutrient losses to surface waters through runoff. However, the authors acknowledge that this practice is not always feasible, because weather can be very unpredictable and producers commonly have a limited time frame in which they can afford the time to apply manure to fields. Results from the N₂O portion of this study do indicate that whenever possible, producers should definitely try to avoid applying manure or fertilizers prior to extreme rainfall events.

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